

REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188	
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1. AGENCY USE ONLY (Leave Blank)		2. REPORT DATE		3. REPORT TYPE AND DATES COVERED Final report, May 1, 1999 – Oct.31, 2004
4. TITLE AND SUBTITLE Hybrid Molecular and Spin-Semiconductor Based research			5. FUNDING NUMBERS DAAD19-99-1-0198	
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9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211			10. SPONSORING / MONITORING AGENCY REPORT NUMBER 39832.25-EL-MUR	
11. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.				
12 a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.			12 b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) This project has three thrusts. Thrust I is entitled “Integrated Bacteriorhodopsin-Based Photodetectors” and its objective is to enable a new class of optoelectronic devices by combining the versatility of genetically engineered biomolecules with the power of microelectronics. Thrust II is entitled “Spintronics and Magnetoelectronics” and its objective is to enable a new class of semiconductor-based spintronic devices. Thrust III is entitled “Molecular Electronics” and its objective is to develop, characterize and model organic/inorganic heterostructures and to identify novel device structures based on these systems. The most important results and publications are summarized in this report.				
14. SUBJECT TERMS			15. NUMBER OF PAGES 6	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OR REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION ON THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL	

Hybrid Molecular and Spin-Semiconductor Based Research

Statement of the problems studied

Thrust I: Integrated Bacteriorhodopsin-Based Photodetectors

The objective of this thrust was to enable a new class of optoelectronic devices by combining the versatility of genetically engineered biomolecules with the power of microelectronics. The unique photoelectric effect and the robust nature of Bacteriorhodopsin (bR) have been utilized to construct novel molecular sensors, modulators, or imaging devices. The light induced photovoltage in the bR is transformed to a photocurrent by a transistor into a useful output. The opto-electronic integrated circuit (OEIC) demonstrated by us opens up the possibilities of realizing more complex optoelectronic devices and circuits with integrated bR as an ultrasensitive, and possibly polarization-dependent, photodetection material for applications in polarization-dependent optical interconnects. Its further integration with semiconductor components will lead to ultra-sensitive sensors with polarization selectivity and built-in preprocessing functions that would have great potential in biological detection, night vision and optical interconnects.

Thrust II: Spintronics and Magnetoelectronics

The objective of this thrust was to enable a new class of semiconductor-based spintronic devices. We pursued this objective at two levels. At the macroscopic level of device synergy, we have demonstrated the monolithic combination of magnetoresistive devices with semiconductor lasers to create a new class of miniaturized magnetic field sensors. At the microscopic level, (1) coherent magnetization rotation and control by ultrashort pulses was demonstrated in half-metallic CrO₂ thin films which are of great interest for spin injection/detection, (2) spin-based light-emitting diodes were demonstrated, (3) optical means were used to coherently control the electronic spins of single and coupled quantum dot structures demonstrating a basic quantum logic device and (4) spintronic device models were developed to study new concepts that couple localized spins to transistor structures.

Thrust III: Molecular Electronics

The objective of this thrust was to develop, characterize and model organic/inorganic heterostructures and to identify novel device structures based on these systems. The theoretical work was focused on the development of a model for current flow through molecules by combining a standard quantum chemistry software with the non-equilibrium Green function (NEGF) formalism for quantum transport. This model is based on the self-consistent field method and new approaches to modeling molecular conduction in the Coulomb blockade regime were also developed. The experimental work funded under this program included (1) application of scanning probe microscopy to characterize bR and other molecular monolayers, (2) development of test bed structures for molecular conductivity (metal/molecule/metal systems), (3) development of chemically and electronically stable semiconductor surface layers, suitable for integration of molecular-scale structures and 4) development and characterization of metal/molecule/semiconductor device structures suitable for electronics, optoelectronics or sensing applications.

Summary of the most important results

Thrust I: Integrated Bacteriorhodopsin-Based Photodetectors

Modeling of the Photoelectric Response of Oriented bR Film: The dynamics of the photochemical cycle of bR has been modeled, and a proposed equivalent circuit was shown to describe the measured bR photoresponse with reasonable agreement.

Measurement of Photoresponse Time of bR: The photovoltaic signal associated with the primary photochemical event in an oriented bacteriorhodopsin film was measured by directly probing the electric field in the bacteriorhodopsin film using an ultrafast electro-optic sampling technique. We showed that the formation of the K state is responsible for the onset of the photovoltage while the decay rate of the photovoltaic signal is due in part to relaxation processes of the protein indirectly coupled or precedent to the formation of the KL state.

bR-FET Monolithically Integrated Photoreceivers: We have fabricated and characterized a bR/gallium arsenide field-effect transistor (FET) monolithically integrated photoreceiver in which the photovoltage developed across the bR is converted to an amplified and useful photo-induced current signal with a responsivity of 3.8 A/W. The schematic and equivalent circuit of the integrated bR/MODFET photoreceiver are shown in Fig. 1(a) and a representative photomicrograph is shown in Fig. 1(b).

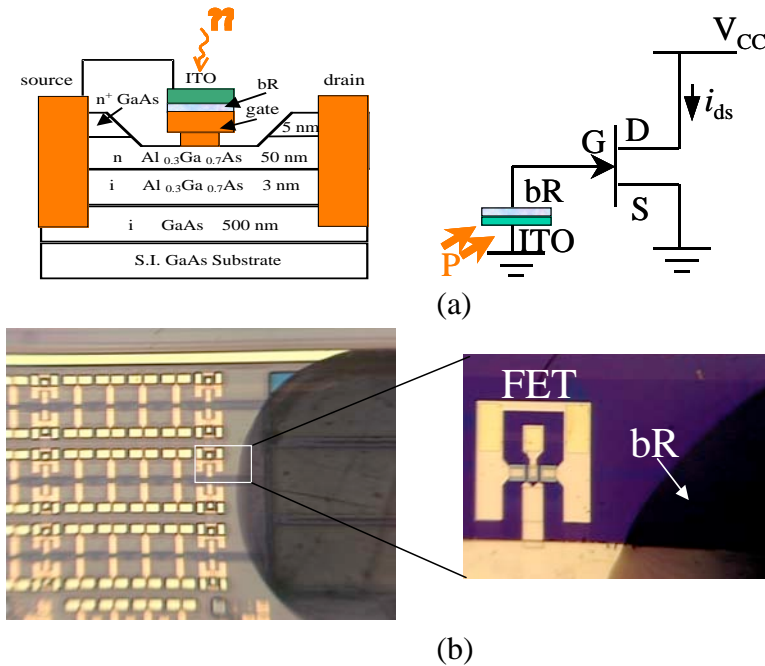


Figure 1: (a) Schematic (left) and equivalent circuit (right) of a monolithically integrated bR/GaAs MODFET photoreceiver structure; (b) photomicrograph of a fabricated chip showing MODFET devices and the selectively deposited oriented and dried bR film.

bR-based phototransceivers: Efforts were also invested to integrate the bR-based detectors vertically with light-emitters such as LED's and VCSEL's, to form a dense array of phototransceivers (Fig. 2) . In this structure, the photocurrent generated by the bR/MODFET photoreceiver were utilized to modulate the output of an integrated light emitters. Such a densely packed two-dimensional array of phototransceivers can detect, process, and transmit information with high sensitivity and efficiency, low-power dissipation, and compact size, and thus would be an attractive component in imaging sensing and information transfer applications. Two techniques were developed to achieve polarization sensitivity of the bR photoelectric response, namely photoinduced anisotropy and chemical bleaching process.

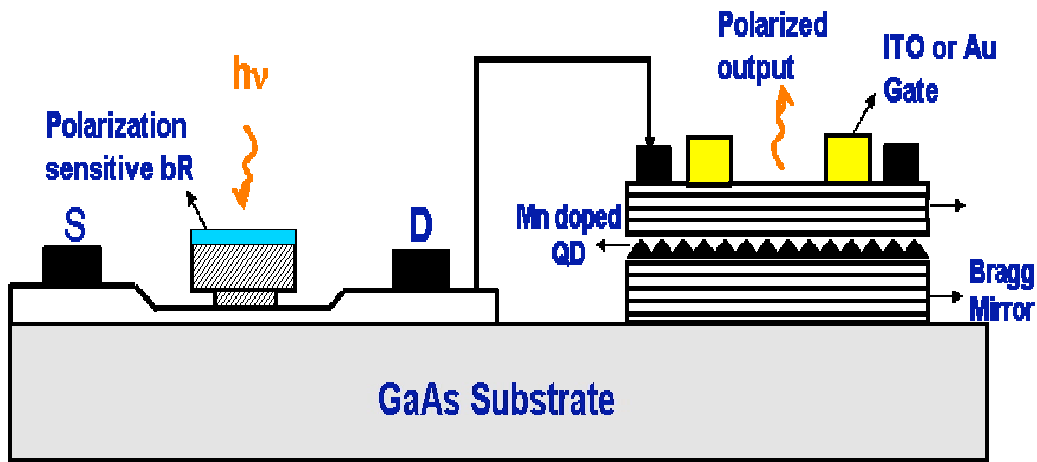


Fig2. Schematic of a bR based Polarized Phototransceiver.

Genetic Optimization of Bacteriorhodopsin for Device Applications: Efficient methods to generate random mutations within the protein were developed, under the assumption that a small number of these mutations will result in a protein that has improved properties relative to the native protein for device applications.

It is important to note that in the integrated photoreceiver, the light induced photovoltage in the bR is transformed to a photocurrent by the transistor and is a useful output. The bR detector can be made smaller in thickness and diameter, whereby the responsivity can be enhanced further. By the very nature of the bR photoresponse, a transient photocurrent, with a fast risetime and a slower decay time, is recorded. The photoreceivers, therefore, cannot be used for constant illumination. However, they would be very useful for dynamic image analysis, motion and edge detection and a variety of other applications.

Thrust II: Spintronics and Magnetoelectronics

Hybrid Magnetic Tunnel Junction/Vertical Cavity Surface Emitting Laser Field Sensor Device: Sensing magnetic fields “wirelessly” is of interest in those conditions where access to the location of the field of interest is awkward, or where such a location is hostile or subject to electromagnetic interference. The circumstances span a wide range of applications, from geological sensing to high-speed readout of magnetic storage media. We have designed and constructed a chipscale magneto-optoelectronic device, composed of a magnetic tunnel junction (MTJ) and a vertical cavity surface emitting semiconductor laser (VCSEL). The hybrid device converts changes of a local magnetic field to an optical signal which in turn may propagate over large distances, either in free space or in an optical waveguide.

Coherent Magnetization Rotation and Control by Ultrashort Optical Pulses in CrO₂ Thin Films: CrO₂ is of particular interest in the field of spintronics due to its half-metallic nature (spin polarization ~96%) which should allow extremely efficient spin injection / detection. In semiconductors, optical techniques have been used recently to create transient populations of nonequilibrium spins for study and control of their transport, phase, and entanglement. By contrast, little such work exists in ferromagnetically ordered metals where the exchange coupling leads to long range order in the ground state of the many-electron system. We have demonstrated that it is possible to employ ultrashort, pulsed laser excitation of a thin ferromagnetic CrO₂ thin film to trigger coherent transient magnetization rotation on a subnanosecond time scale, where the macroscopic photoexcited volume acts as a single precessing magnetic domain. A second feature of the present work is that, by applying the photoexcitation in pairs of time sequenced pump pulses, the phase of the transient precession of the magnetization vector can be controlled, constructively or destructively, depending on the temporal separation between the two impulses of excitation. This mechanism opens the door to modulation of the collective, long range magnetic order via one electron/spin excitation by photons.

Magnetization Reversal of CrO₂ Nanomagnet Arrays: Given both fundamental questions as well as applied potential about magnetic materials on a submicron scale (referred here as ‘nanomagnets’), we undertook an initial study of the magnetic characteristics of patterned submicron nanomagnet arrays of CrO₂ fabricated by electron-beam lithography and reactive ion etching. We showed that the magnetic response can show many facets of behavior with only moderate changes in their geometry and different parameters of size, but it is clear that considerably more detailed and systematic future studies are required.

Optically induced quantum mechanically entangled states and readout: InAs self-assembled quantum dots (SAQD's) have small optical dipoles. This creates a major challenge for optics experiments that rely on the optical dipole for coherent optical control techniques and detection. However, we developed a processing approach that increased the net oscillator strength and which then enabled us to demonstrate optically induced entanglement of two different exciton Bloch vectors. Using this result and our demonstration of the biexciton state, we can now demonstrate a full conditional quantum controlled NOT gate as we published in Science using GaAs. This work demonstrated that the complex manybody physics of higher dimensional systems did not intrude in

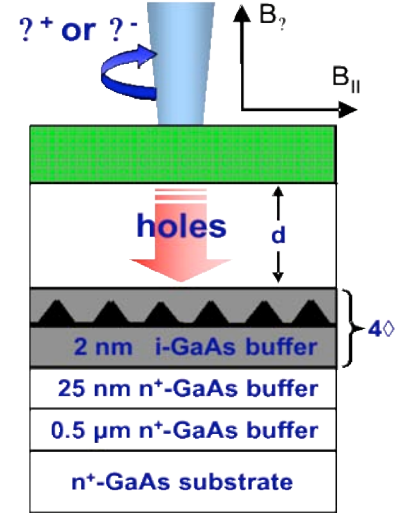
quantum dots, and that quantum dots, with adequate 3-d confinement, behave for the most part as simple atomic systems.

Coherent Optical Control of Electron Spin: The previous work was based on pseudo-spin rather than the electron spin which required developing quantum dots with one additional electron. For this work, experiments were performed with interface fluctuation quantum dots (IFQDs) in GaAs where we have demonstrated the first observation in any system of spontaneously generated coherence through quantum beat experiments.

Spin-polarized lasers: Spin-polarized light sources utilize ferromagnetic semiconductors to inject spin-polarized carriers and ascribe a net circular polarization to the emission upon radiative recombination of spin-polarized holes (electrons) with unpolarized electrons (holes). We have epitaxially grown, fabricated, and characterized spin-LEDs with (Ga,Mn)As spin-aligner layers and $\text{In}_{0.4}\text{Ga}_{0.6}\text{As}$ quantum dot active regions exhibiting large polarization efficiencies of 30% and high-temperature operation $T < 125$ K. We have developed a technique for the self-organized epitaxy of Mn-doped InAs dilute magnetic quantum dots (DMQDs) exhibit room-temperature ferromagnetism. Using a spin-aligner embedded with InAs:Mn DMQD nano-magnets, we have demonstrated high-temperature operation ($T < 180$ K) of spin-LEDs injecting spin-polarized holes into $\text{In}_{0.4}\text{Ga}_{0.6}\text{As}$ QDs. The concept of spin injection from ferromagnetic to nonmagnetic semiconductors may also be applied to lasers.

We have studied spin injection and transport in a (Ga,Mn)As-based spin-polarized vertical-cavity surface-emitting laser (spin-VCSEL) with an $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}/\text{GaAs}$ quantum well active region. A degree of circular polarization of 4.6% was achieved at 80 K, verifying spin injection and transport across a distance of $\sim 0.25 \mu\text{m}$. Controlled modulation of the polarization was demonstrated using applied magnetic fields over a temperature range of 80–105 K. Together with electric-field control of ferromagnetism and high- T_C aligner layers, the spin-VCSEL should find applications in cryptography, reconfigurable optical networks, advanced optical switches and modulators, and light-wave networks with enhanced bandwidth and security.

Development of spintronic device models and new device concepts: Research on semiconductor spintronics for over a decade has been motivated by a variety of novel devices including both conventional and quantum logic. Conventional device concepts are typically based on delocalized band electrons while quantum device concepts are based on bound electrons and nuclei with localized spins. We have (1) developed models for conventional spintronic devices including spin dephasing processes (which are commonly ignored) and (2) developed new device concepts based on the interaction of delocalized electrons and localized spins that may be generally useful in a wide range of spintronic devices, including both conventional and quantum computing. This work is being continued beyond the present project.



Thrust III: Molecular Electronics

Scanning probe microscopy of bR was used to establish a number of results that are especially relevant for future use of bR in bioelectronic applications.

- A demonstration that a measurable photovoltage can be obtained from wild type bR when deposited on substrates chosen for their compatibility with the semiconductor industry (non-aqueous environment). The time dependence of the photovoltage was measured and found to depend on humidity. An equivalent circuit and analytical model for the photovoltage capable of explaining the observed behavior was developed.
- The future use of bR for device applications will almost certainly require the positioning or patterning of wild type bR at the sub-micron length scale. Using electrodeposition techniques and microfluidics technology, we have developed techniques that allow the deposition of bR patches onto substrates in a patterned way.
- All genetically modified bR mutations studied (~ 14 in number) have shown a physically degraded condition when compared to wild type bR. The deterioration observed in bR formed by genetic mutations exhibit either i) large holes permeating the purple membrane bilipid membrane patches or ii) a complete loss of structural integrity of the deposited membrane. This finding raises the question whether genetically modified bR is compatible for use in future bioelectronic devices.

Use of higher harmonics in scanning probe microscopy: During the course of the above work, a systematic study was performed on the higher harmonic content in the vibration spectrum of scanning force cantilevers. The presence of these higher harmonics was originally noticed when studying bR samples in a scanning probe microscope. For a scanning force cantilever oscillating in the attractive regime we have shown that (a) the magnitudes of the higher harmonic signals in the vibration spectrum are directly correlated to the local van der Waals forces, (b) the higher harmonic resonances are much sharper than the fundamental harmonic, and (c) the frequency shifts in the n^{th} harmonic are n times more sensitive to van der Waals forces compared to the primary harmonic. Consequently contrasts in the amplitudes of the higher harmonics over a scanned sample reflect changes in specific chemical composition. These results suggest that a systematic measurement of higher harmonics in the attractive regime can lead to highly sensitive scanning probe techniques to map the chemical composition over heterogeneous samples.

Experimental studies of electrostatic surface potentials of various organic monolayers on Au and GaAs substrates. The goal of this study was to understand the effects of molecular dipoles and charge transfer between the molecule and substrate. The results from this study provide insights into band line-ups and charge dipoles in molecular systems, which are important parameters for modeling efforts in molecular conduction, molecular-based memories, and potential sensing applications. Surface potentials of 10 to 200 mV were observed in the self-assembled monolayers (SAMs) we studied. In general, molecules with a net dipole exhibited surface potentials consistent with the dipole of the isolated molecule, while molecules without a net dipole exhibited negligible surface potentials. From this, we concluded that the primary contribution to surface potential was the intrinsic dipole of the molecule, and that there is very little net charge transfer from the substrate to the molecule (less than 0.05 electrons per molecule).

Development of a few-molecule test bed for molecular conduction. The test bed consists of a break junction, i.e. a nanoscale gap in a thin Au film, formed by passing current through the lithographically defined Au structure. Organic molecules with bonding

groups at each end are then deposited across the break junction by immersion in a solution containing the desired molecule. The measured current-voltage relationships for samples containing short aromatic molecules indicate that the structure allows characterization of a small number of molecules -- ideally a single molecule. The lack of a conduction gap (at low voltages) indicates that relatively strong coupling has been achieved between the contacts and the molecule, i.e. that there is low contact resistance. This structure has been used to study the conductivity properties of several small organic molecules, including the temperature dependence of the conductance. It has also been applied to measure conduction through DNA molecules, which could provide a means to have direct electronic readout of biosensing devices and is being continued under separate funding (NASA).

Development of chemically and electronically stable semiconductor surface layers. In typical semiconductor device or contact structures, it is difficult to make high quality contact at the nanoscale, due to surface states, oxidation and band bending over dimensions much larger than molecular elements. In addition, many semiconductors, including n-type GaAs exhibit “surface Fermi level pinning”, i.e. the measured Schottky barrier height is roughly constant as the metal work function is varied. We have developed chemically stable cap layers by using thin (2-3 nm) thick layers of low-temperature-grown (LTG) GaAs, i.e. GaAs grown at lower than normal substrate temperatures in a molecular beam epitaxy system. This material has approximately 1% excess As, resulting in a very low minority carrier lifetime, and consequently a significantly lower surface oxidation rate. We have demonstrated that Schottky diodes formed on thin LTG:GaAs cap layers on n:GaAs substrates exhibit Schottky barrier heights that vary with the metal work function. This indicates that the surface Fermi level is not pinned in this case, making the surface useful for integration of nanostructures.

Semiconductor-based molecular electronics: Most molecular electronic devices reported to date have been metal/molecule/metal structures. The use of a semiconductor contact for molecular devices allows the use of direct covalent bonds, which should provide more stable bonds (both mechanically and electrically). In addition, the integration of molecular elements with semiconductors can enable hybrid molecule/semiconductor electronics, which could provide new approaches for high density memory and integrated chemical sensing/signal processing platforms. We have (1) developed and characterized metal/semiconductor interfaces with unpinned Fermi levels which provide a means to achieve the controlled surface electronic properties required for nanoscale devices, (2) demonstrated metal/molecule/semiconductor device structures in which the conductivity is higher than comparable metal/semiconductor structures and (3) investigated the dependence on molecular properties including charge dipoles and molecular length. The experimental results are being correlated with our theoretical models.

Theoretical models for molecular electronic devices: Over the last few decades, quantum chemists have developed very sophisticated models that are used to predict the electronic structure of isolated molecules in equilibrium. We coupled one such quantum chemistry software (Gaussian) with the non-equilibrium Green function (NEGF) formalism) to develop a powerful new approach for problems involving current flow in device configurations. However, this approach relies on the self-consistent field method and we have extended it to include Coulomb blockade effects in devices with weakly coupled contacts. This work is being continued beyond the present project.

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Ph. D. degrees: G. Ju, Q. Zhang (expected Spring 2005)

Report of Inventions

“Magneto-optoelectronic switch and sensor” , A.V. Nurmikko, pending (U.S. Pat Office filing # 20050018725)

Awards and Honors

1. P. Bhattacharya, 2002, Nick Holonyak, Jr. Award, Optical Society of America
2. P. Bhattacharya, 2003, Quantum Devices Award, International Symposium on Compound Semiconductors
3. S. Datta, 2001, SRC Technical Excellence Award (with M.S. Lundstrom)
4. S. Datta, 2002, IEEE Cledo Brunetti Award (with M.S. Lundstrom)
5. R. Birge, 2002, 3M Award of Canada in Physical Chemistry.

Technology Transitions

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